ANL/CFCT-20/19

Advanced Moderation Module

Advanced Moderation Module for High-Temperature Micro-Reactor Applications

Chemical and Fuel Cycle Technologies Division
Nuclear Science and Engineering Division

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prepared by
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EXECUTIVE ABSTRACT

Development and deployment of micro-reactors that provide competitive efficiency, prevailing compactness, and inherent safety is an immediate focus of the U.S. nuclear industry. For thermal neutron micro-reactors operating at elevated temperature for optimized efficiency, such as micro molten-salt reactors (MSRs), heat-pipe reactors, and very-high temperature reactors (VHTRs), high-performance moderator based on metal hydride can enhance the neutron economy and therefore achieve reduced weight and enhanced portability. As unclad metal hydride inevitably decomposes at elevated temperature, an enclosure is required for hydride moderator to deliver desired performance at elevated temperatures. Conventional moderator enclosure solutions based on high-temperature alloys introduce extraneous neutron penalty into the micro-reactor, affecting the neutronic benefits provided by the hydride moderator. Additionally, the compatibility between the high-temperature alloy enclosure and the micro-reactor matrix is also a potential issue.

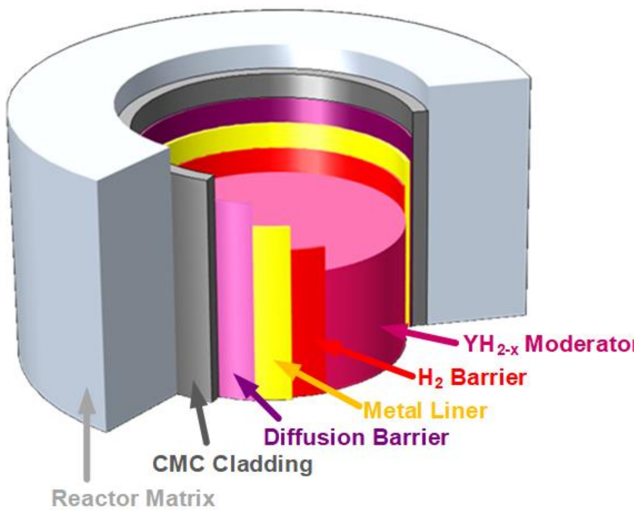

Figure A1. Schematic description of the AMM technology

In this report, we disclose an Advanced Moderator Module (AMM) concept enabled by an innovative enclosure solution combining the advantages of refractory metals, ceramic matrix composites (CMCs), and advanced coating technology to serve as hydrogen permeation barrier up to very-high temperatures. A schematic description of the AMM structure is illustrated in Figure A1. The AMM contains a moderating material core made of metal hydride with high thermal stability, such as YH_{2-x}. The hydride core is enclosed by an H_2 barrier layer coated on a ductile refractory metal liner to minimize hydrogen loss during high-temperature operation. A ceramic matrix composite (CMC) cladding is adopted to provide further structural strength, especially during power transients. Between the CMC cladding and metal liner, an extra diffusion barrier coating is inserted to suppress the chemical interaction at elevated temperatures. Hence, based on a series of innovative material solutions, the AMM is capable of containing the metal hydride core at elevated temperature (>900^{o}C) inside coated and lined CMC envelop with negligible hydrogen loss throughout the microreactor lifetime.

The benefit of the AMM technology was assessed based on a comprehensive multi-stage reactor-physics analysis. Advanced moderation based on hydride metals (such as YH_{2-x}) enables reaching optimum moderation with higher fuel content than traditional VHTR technology, which is required to design compact micro-reactor cores targeting long-life operation. The AMM enclosure technology provides lower thermal neutron absorption rates than traditional solutions (based on stainless steel or Mo-based alloy such as TZM), which reduces the fissile enrichment requirements by 6-8% on a TRISO-fueled design based on the EMPIRE core. Finally, combining the hydride moderator with neutron transparent enclosure solutions provides significant potential to boost neutronics performance of micro-reactors in terms of increased core lifetime or reduced size and weight by 30-50% on a micro-reactor based on the Holos Quad technology.

The progress and plans for on-going development and demonstration efforts are also discussed in this report. The current demonstration of AMM is focused on the hydrogen diffusion barrier demonstration under thermal cycling, while radiation tolerance demonstration is planned. The next step of the demonstration plan will be the assembly of miniature AMM for high-temperature testing, which can be used as a prototype for future pilot scale demonstration.

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1 Introduction

Development of micro-reactors that can be easily transported to remote areas for civilian or military applications is an immediate focus of U.S. nuclear industry and therefore is being pursued by multiple reactor vendors (e.g. HolosGen LLC, Westinghouse, Ultra Safe Nuclear Corporation, URENCO, etc.) and actively supported by DOE-NE, DOE-ARPA-E and by the Department Of Defense (DoD). Very-high-temperature gas-cooled (VHTR) and heat-pipe cooled reactor technologies are regarded as promising concepts for micro-reactors applications since they enable efficient Brayton cycle and display beneficial inherent safety features. Their excellent efficiency originates from their unprecedented high operating temperatures.

High-temperature core concepts traditionally rely on graphite to moderate neutrons since it can sustain high temperatures and displays very low absorption cross-sections. However, it is much less efficient at slowing-down neutrons when compared with a hydrogen-based material [1]. The direct consequence will be to require a high carbon to U-235 ratio in the core. In micro-reactors, a significant amount of low-enriched uranium is needed to be able to sustain several years of operation at high power. Consequently, a large volume and weight of graphite is required in micro-reactors to avoid derating the core power. This directly limits the performance expected for graphite-moderated high-temperature micro-reactors that requires low mass and volume for transportability, and relatively large power output for several years of operation. In addition, there can be significant safety concerns of having a very under-moderated VHTR micro-reactor core when considering accident scenarios associated with water insertion due to flooding. Those potential scenarios may need to be considered for micro-reactors as they can be initiated during reactor transportation (the core sinks into a lake) or during flooding of the installed core in a trench.

Different advanced moderator concepts are currently being investigated in the U.S. for micro-reactor applications with the objective to achieve:

- Increased core compactness (at a given level of uranium enrichment) to reduce size and weight of the core and shielding required for ease of transportation;
- Applicable to high-temperature environment (>700^{o}C and up to 1000^{o}C) to enable improved thermal efficiency and economic targets;
- Improved inherent safety by reducing the reactivity insertion during water flooding scenarios.

This report describes the Advanced Moderator Module (AMM) technology to provide high-temperature core concepts with a boost in moderation. The AMM is not intended at replacing the graphite matrix or nonconventional moderator matrix, but at being placed within the matrix to reduce the amount of graphite needed to reach the optimum for moderation. The AMM is based on a novel material solution that contains the hydrogen, at elevated temperature (>900^{o}C), inside a coated and lined SiC composite envelop. A literature review is presented in Section 2 to provide an overview of the available advanced moderator technologies and their limitations. The basic AMM concept is described in Section 3 together with some demonstrations of the feasibility of this technology. The neutronic benefits of using this technology is quantified in Section 4. Finally, ongoing and planned experimental efforts to demonstrate performance of AMM are described in Section 5.

2 Literature Review on Advanced Moderator Materials

For high-temperature thermal-neutron reactor applications (e.g., micro-reactors), graphite is the most mature selection of available moderators. To reach better moderator performance at high temperatures, hydride- or beryllium-based moderators need to be involved. For beryllium-based moderators, including BeO [2] and metallic Be [3], aside from the fast neutron induced poisoning issue [4][5], dimensional changes and disintegration issues must be mitigated [6–13]. Recently, a BeO/MgO composite solution has been investigated, where compromise in moderation performance [14] is expected. Other moderator materials such as SiC [15,16] may not provide advantageous moderation performance compared to graphite, but have the potential to improve other properties with acceptable compromise in moderation performance.

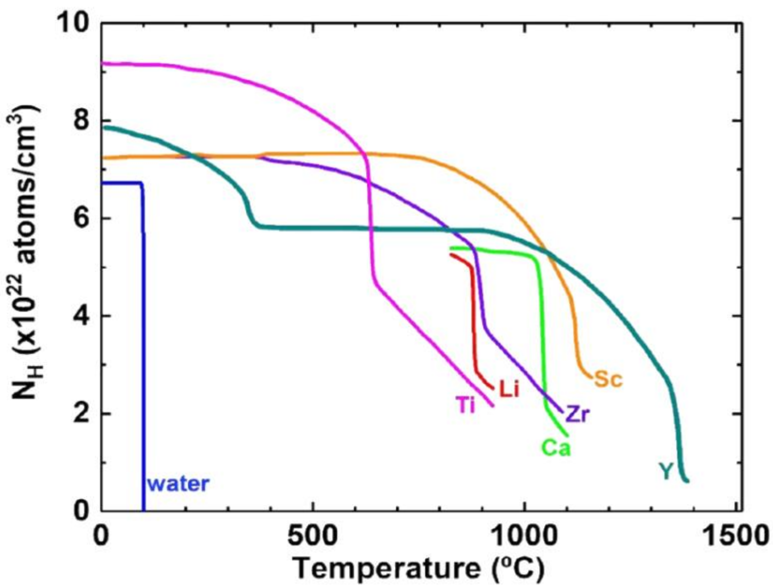


Figure 1 Hydrogen density of different metal hydrides at a constant 1 atm hydrogen partial pressure.

Hydrides of other elements can also be considered as they bring a high density of hydrogen, examples are covalent hydrides, saline hydride, and metallic hydrides. The targeted properties of the hydride are a high hydrogen concentration to enable high neutron moderation performance, low neutron absorption cross-section, sustain high temperature (potentially beyond 900-1000°C) with low hydrogen dissociation temperature at acceptable low partial pressure, and acceptable material properties (high thermal conductivity and ductility, high melting point). If a reasonably low partial pressure (~1 atm) is needed, either calcium hydride (liquid phase or liquid-solid mixture) [17], yttrium hydride (solid phase) [18], or some lanthanide hydrides can be adopted (see Figure 1 , which summarizes data in Ref. [1]). If higher hydrogen partial pressure in the moderator enclosure is acceptable, zirconium and titanium hydrides may also be used. Yttrium hydride seems to be a promising candidate for high-temperature micro-reactor applications given its higher dissociation temperature and stable solid phase.

For high- temperature hydrides (e.g., YH_2), the hydrogen partial pressure is ~1 atm at ~1000°C [19]. Thus, the hydride moderator must be encapsulated. Hydrogen permeates most common structure materials, especially at elevated temperatures. Therefore, the encapsulation approach is expected to involve hydrogen-impermeable materials to maintain adequate hydrogen partial pressure throughout the entire reactor lifetime. However, beyond 900°C, almost all metallic materials have significant hydrogen permeability due to activated diffusion kinetics [1]. Although monolithic ceramic materials provide sufficiently low hydrogen permeability at this temperature [20,21], maintaining an internal hydrogen partial pressure may lead to unfavored tensile hoop stress and subsequent failure risks for monolithic ceramics, especially during power transients. As part of the Aircraft Nuclear Propulsion (ANP) program, a complex solution was developed to encapsulate yttrium hydride for high-temperature application [22]. Cr-doped yttrium hydride was coated by cold-sprayed Cr to enhance its compatibility with FeCrAl alloy cladding. The hydrogen permeation was suppressed by an Al_2O_3 layer formed on the FeCrAl surface. However, once hydrogen partial pressure is significant, the Al_2O_3 can be reduced by hydrogen leading to prominent loss of hydrogen at >950°C [18,22]. Therefore, it is important to use ceramics that do not react with hydrogen at the operating temperatures. On the other hand, ceramic matrix composite materials (CMCs), such as SiC/SiC, C/SiC and C/C, have been developed to provide considerable macroscopic tensile ductility [23]. Such CMCs still suffer from the microcrack formation (channels for H_2 leaking) at a relatively low tensile stress [24]. Meanwhile, refractory metals are capable of delivering satisfactory mechanical properties and structural integrity at high temperatures. The hydrogen permeation issue of the refractory metals can also be relieved by adopting sufficient wall-thickness and a barrier coating. Therefore, moderator cans made of refractory metals such as TZM (a titanium-zirconium-molybdenum alloy) [25] are being considered for micro-reactors. However, the use of refractory metals as the main structure introduces significant neutron absorption compared to SiC (as discussed in Section 4). Additionally, the thermomechanical and chemical compatibility between refractory metals and reactor matrix may also be a potential issue.

3 AMM Technology Description

The Advanced Moderator Module (AMM) technology [26] is designed to provide high-temperature core concepts with a boost in moderation. This is made possible by enclosing yttrium hydride metal inside a ceramic matrix – using an advanced coating technique – that serves as a hydrogen permeation barrier up to very high temperatures. The different components of the AMM technology are described in Figure 2:

(1) YH_{2-x} metal hydride moderator that provides excellent hydrogen density at high temperature to benefit neutronics.

(2) Thin refractory metal liner such as Nb or Mo that provides a ductile and radiation tolerant substrate for advanced coatings while averting excessive neutron absorption.

(3) Standalone ceramic or ceramic-metal coating with low pin hole defect densities prepared by atomic layer deposition (ALD) to provide exceptional hydrogen barrier performance. Other surface modification methods such as chemical vapor deposition (CVD), electrodeposition, and reaction bond coating technologies may also be used for developing such barrier layers with proper engineering optimizations.

(4) Ceramic matrix composite such as SiC/SiC based cladding that provides high-temperature mechanical strength without introducing an extraneous neutron penalty.

(5) Standalone Ceramic or ceramic-metal coating with low pin hole defect densities prepared by ALD, CVD, electrodeposition, physical vapor deposition (PVD), reaction coating, etc. to provide exceptional diffusion barrier between refractory metal liner and CMC.

(6) Optional: a plenum region can be added to the hydride moderator enclosure as a buffer area. The plenum will allow reversible reduction of H/M ratio (i.e., hydrogen density or moderation efficiency) when the temperature is high. This can work as an extra negative feedback mechanism of reactivity to enhance the safety of the reactor (see Figure 3).

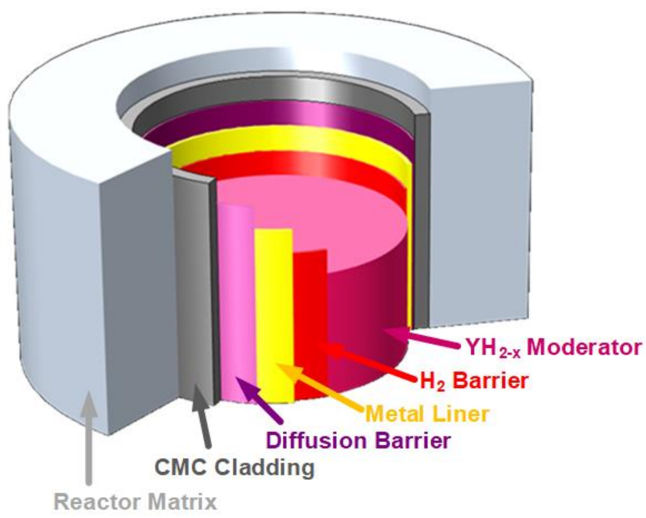


Figure 2. Schematic description of the AMM technology.

Thus, a relatively thin refractory liner with both inner and outer surfaces coated with barrier layers, externally supported by CMC cladding will be used to encapsulate the high-temperature hydride moderator. The inner surface of the refractory liner is coated with an advanced coating containing a

H_2-impermeable material to keep all the hydrogen inside the enclosure and prevent embrittlement of the liner material that result from hydrogen migration in the liner. Meanwhile, the outer liner surface is coated with a diffusion barrier that suppresses the interaction between CMC and refractory metal. This suppression helps in slowing down the possible high diffusion of Si within the metal refractory liner that results in complete consumption of the liner to a more brittle intermetallic compound unable to withstand thermal variations, especially at the high application temperatures [27]. For this AMM design, ALD is the ideal means to apply these advanced coatings, due to its very sensitive thickness control and unique conformal coating ability over long surface areas [28], in addition to its unique ability to produce a layer with very low pin hole defect density . Other coating techniques, such as CVD and reaction coating, may also be used in certain applications with engineering optimization. The SiC/SiC envelope is bonded to the liner's outside surface for sufficient mechanical support to enclose H_2.

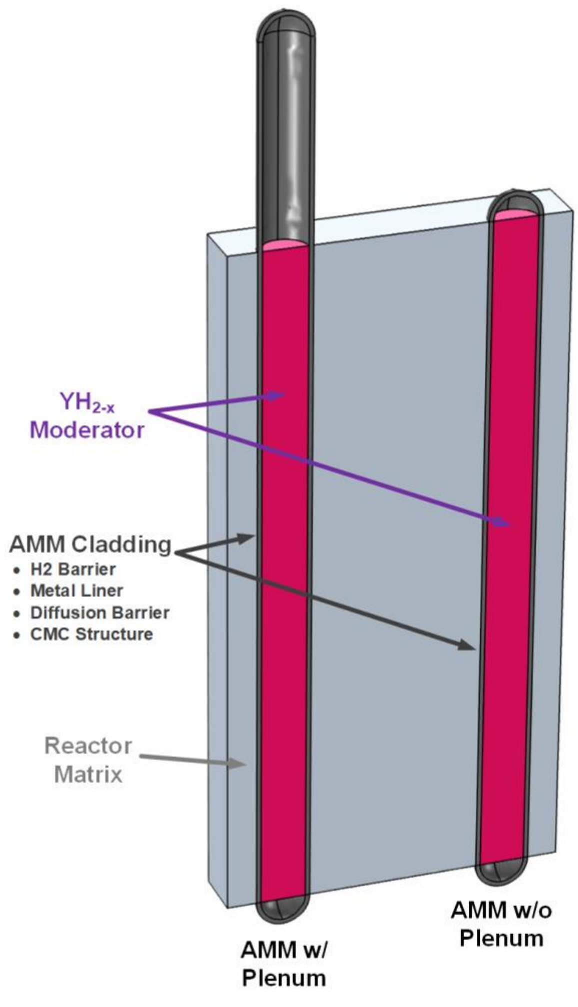


Figure 3. AMM technology with Plenum description.

Additionally, different end-cap solutions can be used for the AMM concept. As shown in Figure 4, the end-cap can either have the similar multi-component structure or consist of thickened metal structure without CMC support.

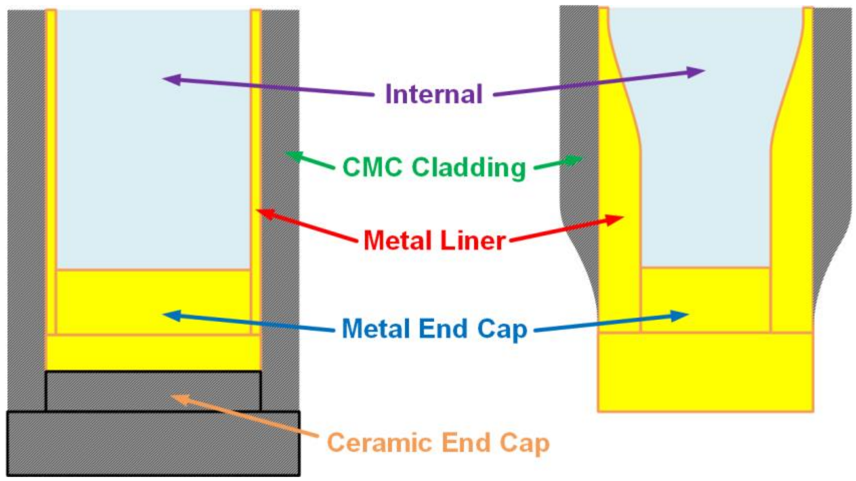


Figure 4. AMM technology with end-cap description.

4 Quantification of Neutronics Benefits to Micro-Reactor Applications

The benefit of AMM technology is assessed with multi-stage reactor-physics analysis. First, the unit cell analysis of the traditional TRISO-fueled reactor is performed to understand the potential benefit of using advanced moderator in a thermal reactor. The AMM is then applied to a thermal-spectrum heat-pipe reactor concept to quantify the benefit in terms of reduced fissile enrichment. Then, full-core design optimization is completed to quantify the gain obtained in terms of core lifetime and weight with the AMM. For all these analyses, the SERPENT code [29] is used for eigenvalue and burnup calculations based on the ENDF/B-VII nuclear data library.

4.1 Infinite lattice analysis

As a first stage, the neutronic impact of the advanced moderator is assessed based on 2-D assembly description with a reflective boundary condition. The proposed assembly design is based on traditional VHTR technology as presented in Figure 5, and contains 19 fuel pins with TRISO packing fraction of 40%. Each fuel compact is being surrounded by six flow channels. A layer of graphite surrounds the assembly to provide additional moderation. The revised assembly geometry is shown in Figure 5 where the assembly is surrounded by AMM pins made of yttrium hydride (YH_2) and using different materials as enclosure solutions. The radius of the YH_2 pin was roughly optimized to reach optimum moderation fraction at 40% packing fraction.

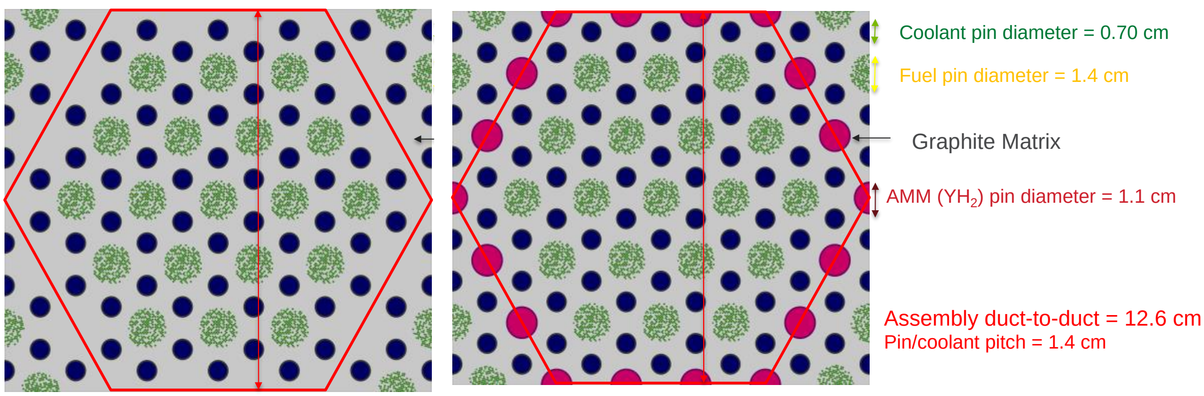


Figure 5. Assembly cell geometry with and without the AMM.

The main results of this analysis are shown in Figure 6 with the neutron multiplication factors (simply, k-infinity) of a TRISO-fueled, graphite-moderated assembly as a function of TRISO fuel packing fraction in a thermal neutron reactor without and with the AMM replacing some graphite at the periphery of the assembly. In this sensitivity analysis, the moderation over the fissile ratio was changed by varying the TRISO packing fraction. In the reference assembly design (no AMM), the k-infinite increases significantly as the packing fraction decreases (black curve in Figure 6) proving that this assembly is very under-moderated: the optimum of moderation is achieved with a packing fraction of ~5-10%.

However, for SMRs and micro-reactors, a low TRISO packing fraction (e.g., ~10%) limits power and operation time. Therefore, SMRs and micro-reactors are typically designed with TRISO packing fraction of 30–40%, or with plain fuel pins with even larger volume fraction, to make the core compact and commercially viable, which is far from the optimum moderator-to-fuel value. Thus, high-performance moderator that slows down neutrons more efficiently than graphite is needed to move the moderator-to-fuel ratio closer to the optimum value under high fuel fraction, and the AMM technology was developed to meet such requirement.

Consequently, additional cases are considered in Figure 6 using yttrium hydride moderation with various enclosure materials. In all cases, the optimum moderator configuration is successfully achieved around 40% packing fraction (shown in Figure 6). Different cladding materials were considered with 0.5mm of SiC with a 0.1mm Nb liner or a 0.4mm cladding of SiC with a 0.1mm Nb liner (reference AMM solution). The TZM alloy based on Mo, and the Nb materials would be strong candidates to clad YH_2 due to their thermo-mechanical properties and capacity to retain H_2 up to high temperatures, especially when associated with advanced coating. However, both Mo and Nb display a relatively large absorption cross-section that significantly reduces the benefit expected from this technology. As shown in Figure 6, the gain in k-infinite is ~2% at 40% packing fraction with Nb, while TZM provides a significant reduction in k-eff (comparing the red, blue and black curves). Eliminating (by using SiC) or at least reducing the layer thickness of Nb (with only Nb liner of 0.1mm) does bring some benefits with gains up to 10% in k-infinite.

Consequently, the AMM technology based on SiC and thin Nb liner surpasses the neutronic performance obtained with enclosure solutions made of refractory metals using Niobium or Molybdenum alloys. Higher assembly k-infinite with large fuel fractions can be used to design denser and longer-life SMRs or micro-reactor cores. These gains in k-infinite will be directly leveraged to reduce the core fissile enrichment, its size and weight (as discussed in the following sections). There is also a direct safety benefit of designing a reactor closer to the optimum moderation with reduction of inserted reactivity during a postulated water flooding scenario, as also shown in Figure 7. The reduced water flooding reactivity obtained can be more easily over-compensated by reactivity control systems.

Finally, it should be observed that the peak k-infinite expected with YH_2 is smaller by 0.15 when compared with the reference design (using graphite). This is explained by the still relatively significant absorption cross-section of yttrium and hydrogen. Larger gains would be expected by employing other hydride metals such as ZrH_2, but those should only be considered at lower operating temperature and will require higher hydrogen partial pressure. The AMM does bring some extra weight to the system since the density of YH_2 is larger than that of graphite, leading to about 10% increase in assembly weight for the proposed configuration. This increase in assembly weight is compensated by the increase in k-infinite achieved when considering the full core optimization by reducing the number of graphite moderators in the core (as discussed in Section 4.3).

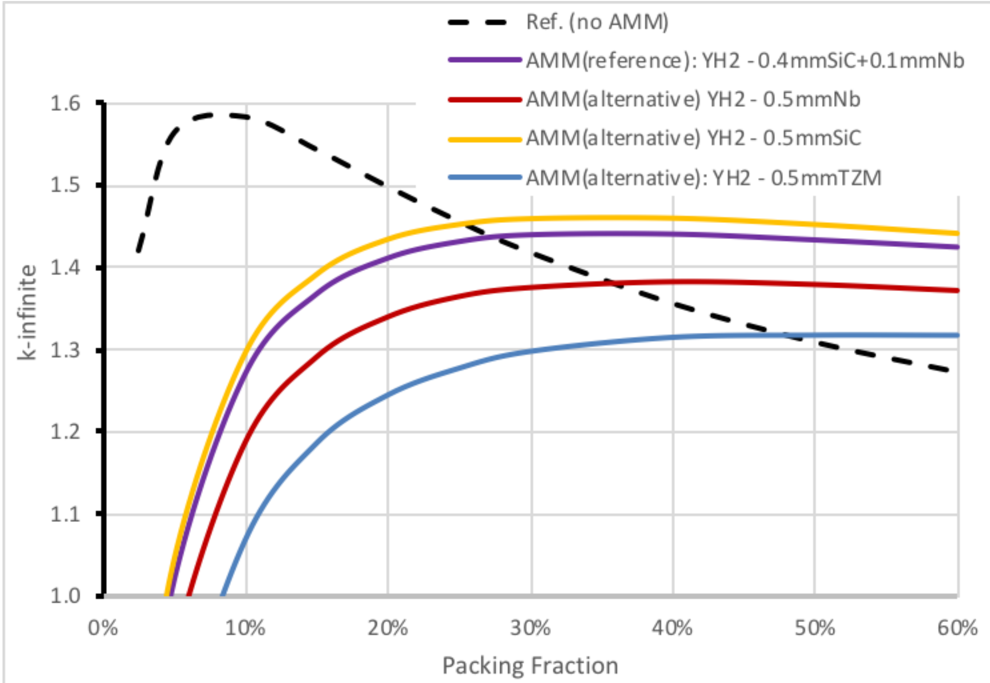


Figure 6. Impact of AMM with different envelope solutions on k-infinite of VHTR assembly design.

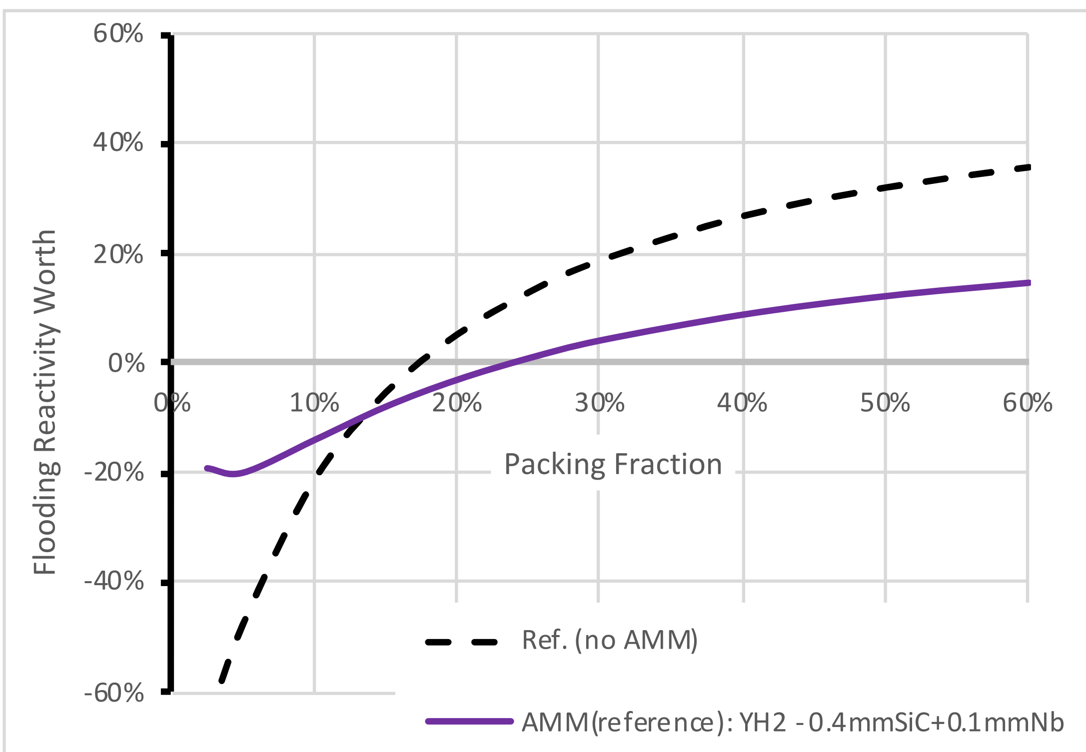


Figure 7. Impact of AMM technology on water flooding reactivity on VHTR assembly design, considering cold water flooding the coolant channels.

4.2 Direct application to heat-pipe micro-reactor design

In the second stage of this analysis, we applied the AMM technology to assess the reduced fissile enrichment in a micro-reactor enabled by the improved neutron transparency of the AMM enclosure. This analysis is based on a TRISO-fueled version of the EMPIRE micro-reactor core developed by LANL [30] that already relies on YH_2 moderator pins enclosed by traditional solutions. This core was modified at ANL using TRISO fuel in a graphite matrix [31] as shown in Figure 8. The reference TRISO-fueled EMPIRE configuration uses YH_2 moderator pins with 0.6mm-thick envelope based on TZM.

The main results from this sensitivity analysis are shown in Table 1. For all core configurations, it was verified that the Moderator Density Coefficient (MDC) is negative and close to 0, meaning that the designs are close to the optimum of moderation. First, it should be noted that the EMPIRE core design would require major changes to enable <19.95 at% enrichment when using TRISO fuel since the neutron flux spectrum is significantly thermalized and high neutron penalty is brought by the TZM material in moderator enclosure and heat-pipe. As expected, the AMM technology reduces neutron penalty from the enclosure of the YH_2 moderator with higher k-eff by ~7,000 pcm (per cent mille) compared to the TZM solution. The AMM enclosure provides a reduced rate of neutron absorption, which is observed with an increased neutron flux in the thermal region, as shown in Figure 9. Without changes in the core design, the estimated reduction expected on U-235 enrichment enabled with the AMM technology is 6 to 8%.

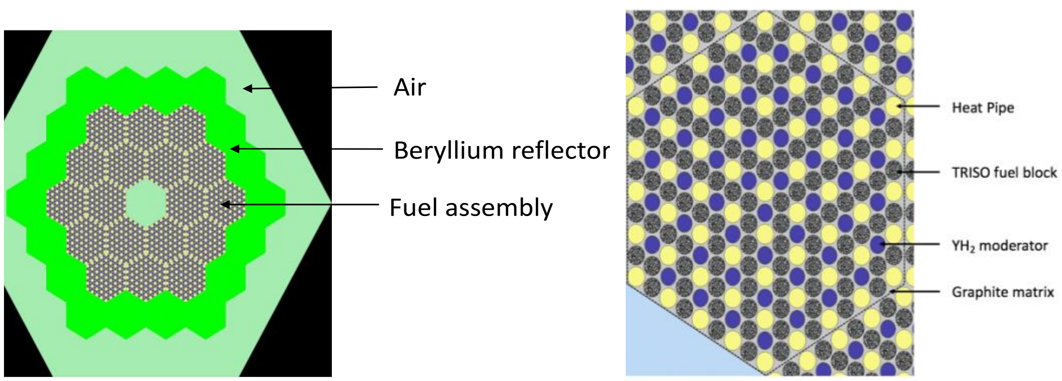


Figure 8. Radial core layout of the TRISO-fueled Heat-Pipe core model and zoom on assembly description.

Table 1. Impact of AMM technology on core eigenvalue and critical enrichment.

Moderator enclosure	k-eff * ± 0.0003	MDC * [pcm**/%] ± 45pcm	Estimated U-235 enrichment needed to reach criticality (k-eff = 1)
TZM	0.88750	-106	33.6%
AMM	0.95165	-57	25.0%

* k-eff and MDC are calculated for U-235 enrichment of 19.95 at.%.

** pcm means per cent mille.

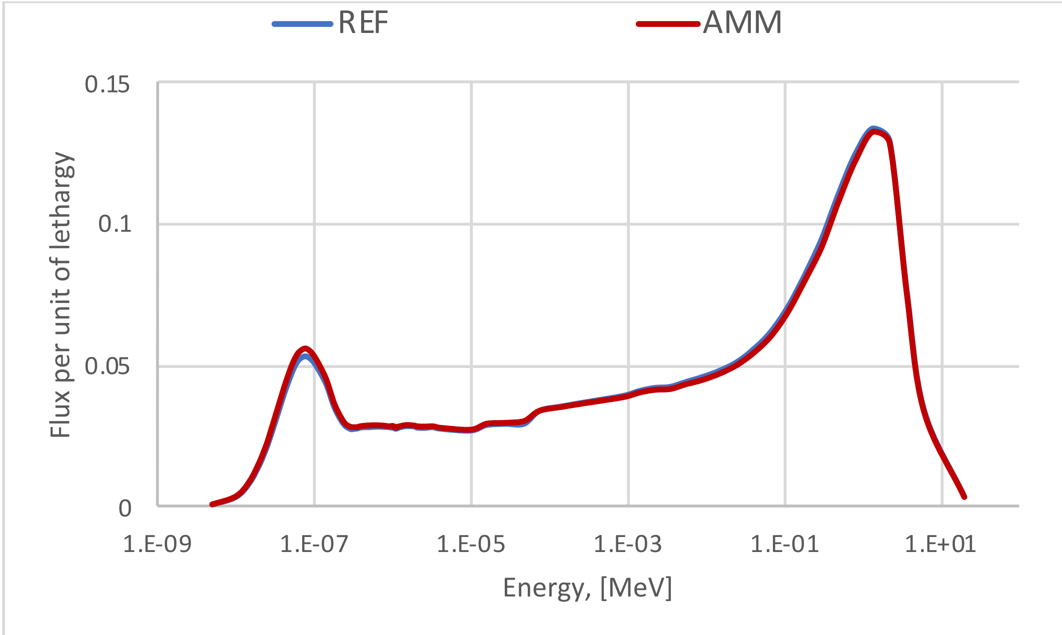


Figure 9. Flux spectrum comparison of the different core configurations (19.95 at.%).

4.3 Application to optimization of a VHTR micro-reactor core

In the final stage of this neutronic analysis, we completed the design optimization performed in [32] to assess the potential of the AMM technology to boost the performance of a defined micro-reactor technology. This study is based on a preliminary version of the 22MW thermal Holos Quad micro-reactor developed by HolosGen LLC, whose core design was optimized at ANL using advanced multi-criteria core optimization methodology [33] under an ARPA-E MEITNER award. In order to provide the comprehensive analysis of the potential of the AMM technology, the same multi-criteria approach based on similar design constraints was applied to the Holos Quad, while enabling AMM rods to replace graphite in between fuel assemblies (as shown in Figure 5).

The Holos Quad micro-reactor, whose radial layout is shown in Figure 10, was optimized targeting increased core lifetime for improved economics and reduced core weight to facilitate transportation. This study quantifies the improved compromise in both competing objectives enabled with the AMM technology. The varying design parameters are listed in Table 2 for both the original optimization performed in [33], and for the updated optimization employing the AMM. The range of some design parameters was adjusted to account for the expected reduction of graphite moderator when relying on the AMM.

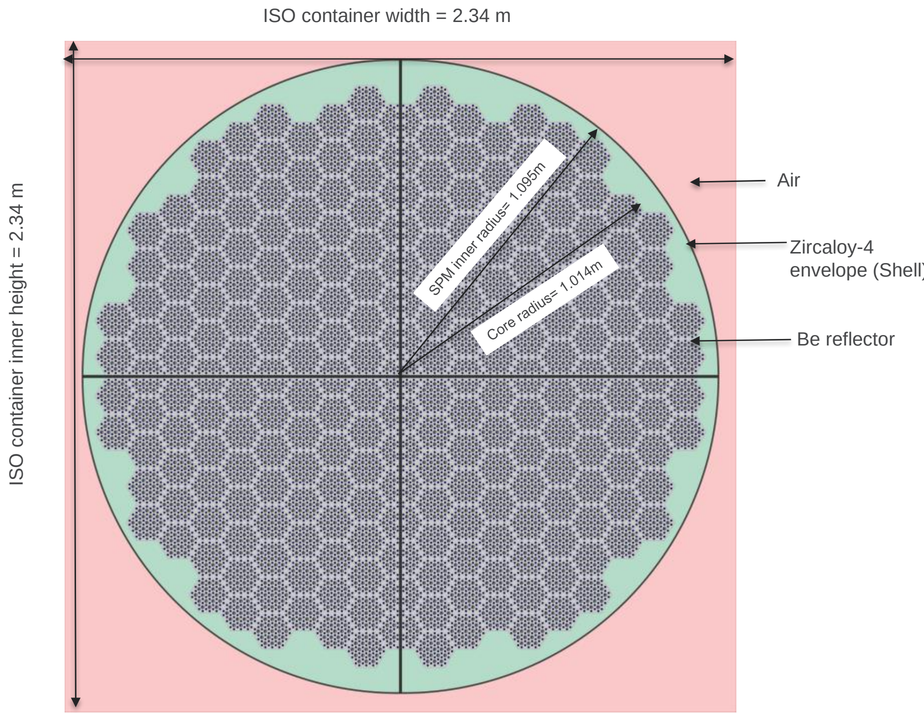


Figure 10. Transversal core layout of Reference Holos Quad configuration [33].

Table 2. Design parameters selected in this core optimization.

Design parameter	*Traditional (No AMM)* [33]	***AMM***
	Lower bound-upper bound	
Height of the driver fuel	200-500 cm	100-400 cm
Height of Axial reflector	5-25 cm	5 cm
TRISO packing fraction	25-40%	30-40%
Number of Assemblies	151-249	109-207
Thickness of radial reflector (estimate of the thickness of reflector between the fuel block furthest away from the center, and the shell)	5/15 cm *(constrained by geometry)*	
Cell pitch	1.4-1.75 cm	1.0-1.5 cm
Inter-assembly graphite layer	0.05-0.4 cm	n/a
Coolant hole radius	0.30-0.38	0.30-0.40
Burnable poison concentration (average, only in graphite block)	10-25 ppm	
AMM outer radius	n/a	0.4-0.99 x (cell pitch – coolant hole radius)

Each design optimization required complete characterization of ~2,000 potential core options to converge to the Pareto Front displayed in Figure 11. On this figure, the blue and red points display the performance of an optimum, fully characterized core option that meets all design requirements. On these Pareto Front graphs, only the best performing cores are displayed to show the optimum compromise achievable between the two competing objectives (core weight and lifetime). This chart shows that the AMM technology can provide reduced core weight (between 30-50%) or increased core lifetime (by at least a factor of two) for the Holos Quad concept. This weight estimate does account for the higher weight of the AMM versus the displaced graphite.

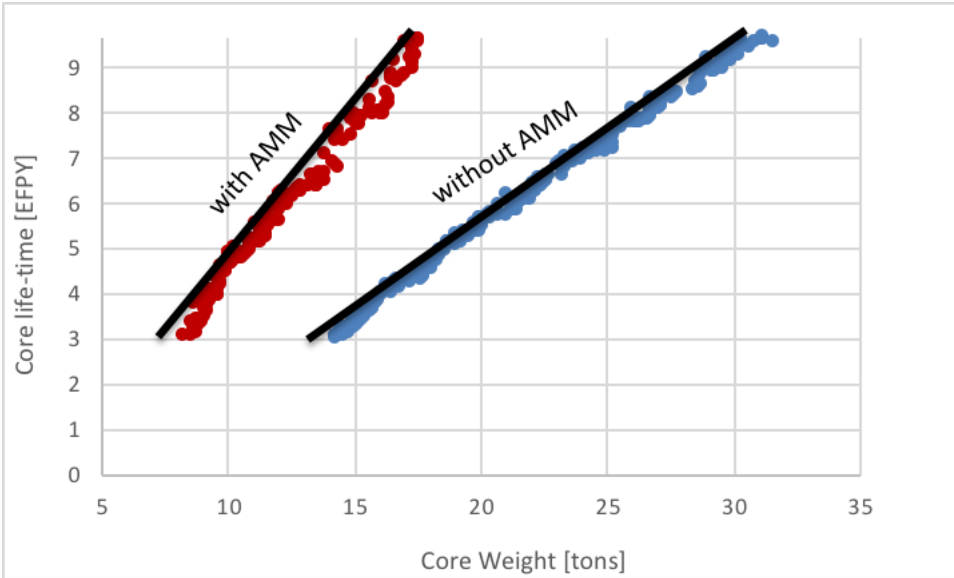


Figure 11. Pareto-frontiers displaying the best achievable core solutions with maximum core lifetime versus minimum core weight that can be obtained with a Very-High Temperature-type thermal neutron reactor with and without AMM.

To conclude, Table 3 summarizes the design characteristics of optimum Holos Quad options targeting eight EFPY lifetime, that were designed with and without the AMM technology. This table shows that applying AMM technology requires complete re-design of the reactor since the optimum solution was determined using very different design options such as number of assemblies, fuel length, cell pitch, etc. The optimum AMM core uses reduced cell-pitch as it does not rely as much on graphite for neutron moderation, however it uses more assemblies to increase fuel content, and a reduced core length to minimize its weight.

Table 3. Main properties from optimum cores targeting 8 EFPY with and without AMM.

	Traditional (No AMM) [33]	***AMM***
Num. assemblies	151	207
Fuel length [cm]	390	300
Cell pitch [pcm]	1.70	1.05
B-10 Burn. Ppoison conc. [ppm]	20	23
TRISO packing frac. [%]	40	40
Be refl. thickness [cm]	7	8
AMM radius [cm]	-	0.66
Weight [tons]	**26.7**	**15.7**
Lifetime [EFPY]	8.3	8.1

4.4 Summary of the benefits of AMM technology to micro-reactor core design

The benefit of the AMM technology was assessed in this section based on a multi-stage reactor-physics analysis. Advanced moderation based on hydride metals (such as YH_2) enables reaching optimum moderation with higher fuel content, which is required for designing compact micro-reactor cores targeting long-life operation. The AMM enclosure technology provides lower thermal neutron absorption rates than traditional solutions (based on stainless steel or Mo-based alloy such as TZM), which reduces the fissile enrichment requirements, while still accommodating high-temperature requirements. Finally, combining the hydride moderator with neutron transparent enclosure solutions provides significant potential to boost the neutronics performance of micro-reactors in terms of increased core lifetime or reduced size and weight.

5 Status of the Technology Demonstration

Many components of the AMM, such as hydride core [25,34] and CMC [35,36], have been extensively studied by multiple programs sponsored by both the federal and private sectors. Their manufacturability and non-nuclear and nuclear performance have been examined. Therefore, the technology demonstration of the AMM needs to be focused on two aspects that have been seldom studied: the performance of H_2 and diffusion barrier coating at high temperature and high radiation conditions; and the assembling of the AMM system.

5.1 Barrier Layer Development and Performance Analysis

5.1.1 Development of H_2 Diffusion barrier Layer

Coating as the most effective method against high temperature H_2 permeation.

At elevated temperatures, the hydrogen permeability of conventional metal-based hydride cladding materials such as stainless steels increase to an unacceptable level. In fact, beyond 900°C [37], all metallic materials have significant hydrogen permeability due to the activated diffusion kinetics [38]. Therefore, it is clear that the barrier material must have good material properties such as a dense microstructure and very low H_2 permeation in order to achieve the theoretical H_2 permeation barrier values. Generally, barriers have been applied as external coatings on existing metallic alloys to prevent hydrogen permeation or hydrogen uptake into the material substrate. Other than coating, material surface treatments (e.g. reaction bond coating) are also designed and pursued to produce an external in-situ scale on the metal or alloy itself that serves as a diffusion barrier. However, such forms of barriers do not provide any flexibility towards stress generated from thermal cycling, which often leads to surface cracks and eventual spallation [39]. Broadly, coating provides more flexibility in its application requirements and can be tuned to generate the most efficient structures to withstand thermal stress and reduce H_2 permeability. Coating also provides the flexibility to develop unique optimized chemistries and control overall barrier thickness (ideal for efficient AMM design) which can withstand this unparalleled high temperature application range of the proposed micro-reactor. Thus, application of the barrier in coating form will be the most effective approach for this AMM design.

Application of barrier coating inside of the refractory metal liner wall.

Due to the need of maintaining a certain partial pressure of hydrogen over the metal hydride in order to keep the necessary H_2/metal ratio, the moderator must be encapsulated hermetically. This entails that the barrier coating will be most effective if applied inside of the refractory metal liner walls, as it will prevent dissolution of H_2 within it. Additionally, it will slow down hydrogen diffusion into the metal liner, because without it, at the reactor operational temperatures excessive amounts of dissolved hydrogen in the liner, may result in matrix hardening and formation of deleterious brittle phases which if formed may lead to crack generation during thermal cycling.

H_2 Barrier coating properties for high temperature environment.

Effective barrier coatings against hydrogen generally possess dense microstructure, with grain sizes in nanocrystalline range with minimal pinhole defect densities [40]. For the barrier coating to be effective at this temperature and compatible with the thin refractory metal liner, it needs good chemical resistance against hydrogen attacks and also able to maintain a low thermal expansion mismatch with the metal liner. These barrier coatings can potentially be fabricated by a variety of techniques, including PVD, CVD, electrodeposition, and various reaction bond coating, with essential engineering optimizations, the metal-ceramic multilayer barrier coating enabled by ALD method [41], which features intrinsic thermal cycling resistance and inherent diffusion suppression, is considered as the most promising candidate for such barrier layers in the AMM concept. Therefore, a series of experiments are being performed and planned to demonstrate the performance of the H_2 and CMC-metal diffusion barriers under micro-reactor conditions. Some preliminary results are reported here. Detailed discussion is presented in Sections 5.1.2 and 5.1.3, where development and testing of various coating designs have been shown to identify the most optimized structure which shows utmost compatibility and thermal stability in the extreme testing environments.

5.1.2 Thermal cycling resistance of advanced ALD multilayer barrier coating

During the micro-reactor operation, the AMM may experience frequent thermal cycling due to either start-shutdown or load following events. Thermal cycling test was then conducted under 4% H_2/96% Ar atmosphere using a tube furnace. Three different types of ALD coatings on Nb substrates were selected for this test: (1) a single-layer Al_2O_3 coating, Fig. 12(a); (2) a ceramic-ceramic alternate Al_2O_3/ZrO_2 multilayer coating, Fig. 12(b); and (3) a metal-ceramic alternate Al_2O_3/W multilayer coating, Fig. 12(c). All specimens experienced four thermal cycles. During each cycle, the specimen temperature linearly ramped up from room temperature to 900°C within 10 hours, maintained at 900°C for 4 hours, and then ramped down to room temperature within 10 hours.

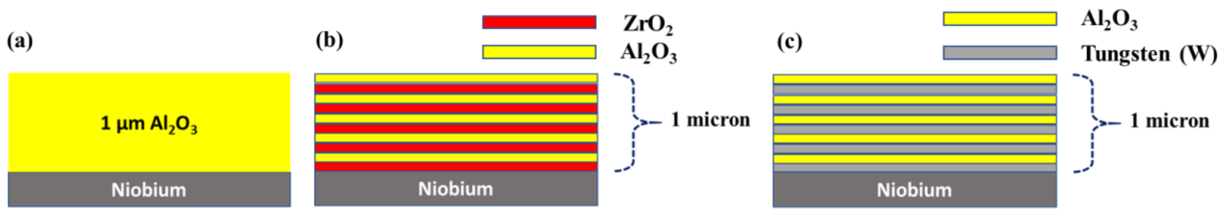


Figure 12. Schematic representation of the ALD barrier coating developed over the Nb coupons: (a) 1 micron thick ALD Al_2O_3; (b) alternate ALD Al_2O_3 and ZrO_2 multilayer to achieve a final thickness of 1 micron; (c) alternate ALD Al_2O_3 and W multilayer to achieve a final thickness of 1 micron.

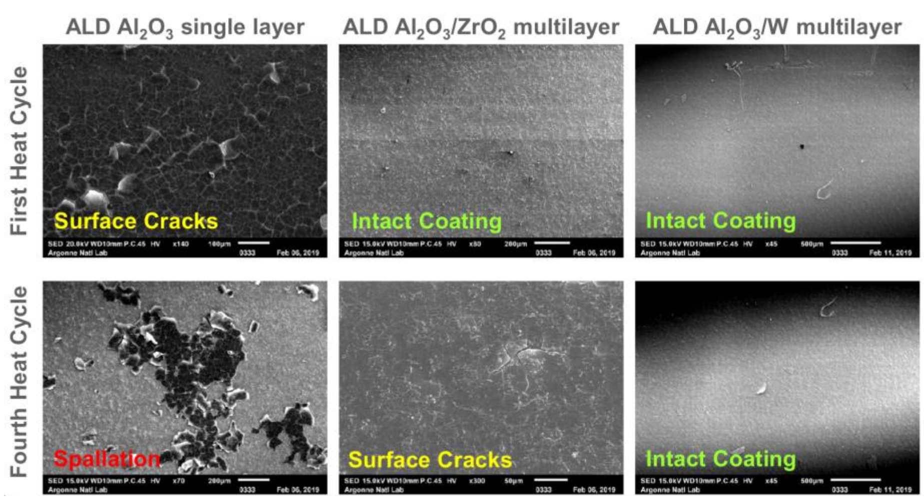
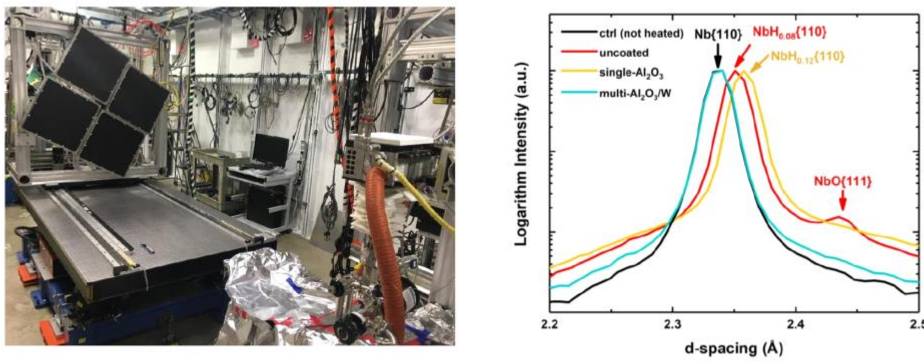


Figure 13. Surface morphology of the thermal cycled specimens after first and fourth thermal cycle.

After thermal cycling, the surface morphology of the specimens was investigated by scanning electron microscope (SEM), as shown in Figure 13. It is obvious that single layer Al_2O_3 coating starts cracking after merely one thermal cycle and suffers severe spallation after four cycles. Meanwhile, the ceramic-ceramic multilayer coating seems to have better thermal cycle resistance, maintaining its integrity after the first cycle. However, after four cycles, surface cracking can still be identified. On the other hand, the metal-ceramic multilayer coating was found to remain intact after four cycles, demonstrating its excellent performance under thermal cycling.



WAXS Setup in APS-1-ID-E

Phase identification based on WAXS

Figure 14. Phase identification of post-test specimen using synchrotron X-ray diffraction.

The post-test specimens were further characterized using synchrotron X-ray diffraction (XRD) at Sector 1-ID-E at Argonne's Advanced Photon Source (APS), as illustrated in Figure 14. As the hydrogen barrier coating is expected to prevent hydrogen from permeating through to hydride the Nb substrate, the {110} diffraction peak of Nb was monitored to assess the degree of hydride formation. It is clear that the single-layer Al_2O_3 barrier cannot protect the Nb substrate from hydrogen attack, as the shift in Nb peak is similar to uncoated Nb exposed to H_2. On the other hand, the Nb sample protected by the metal-ceramic multilayer barrier is the same as the control sample, indicating very effective protection to H_2 permeation. Additionally, it is noticeable that uncoated Nb was also slightly oxidized by the impurity in 4% H_2/96% Ar gas, which was not distinguishable in both single-layer and multilayer coated Nb specimens.

These experiments provide preliminary proof-of-concept of multilayer barrier coating's advantage in relation to thermal resistance. It is worth mentioning that a single-layer coating may also be optimized to provide acceptable thermal resistance through some engineering optimization, such as addition of buffer layer or post-coating treatment.

5.1.3 Hydrogen permeation suppression performance

The hydrogen permeability of the H_2 permeation barrier is a key performance criterion of the AMM. Therefore, a differential pressure system was established to measure the hydrogen permeability quantitatively. The design of the device is illustrated in Figure 15. The device can be divided into two sections: the hydrogen source section and the permeation measurement section. Pressurized 4% H_2/96% Ar gas was used as the hydrogen source, while the permeation measurement section is kept under ultra-high vacuum (UHV) to enable the particle pressure measurement based on a residual gas analyzer (RGA) mass spectrum. The two sections are divided by a disk specimen located in a tube furnace. During the measurement, the specimen was heated so that the permeated hydrogen quantity can be measured by the RGA.

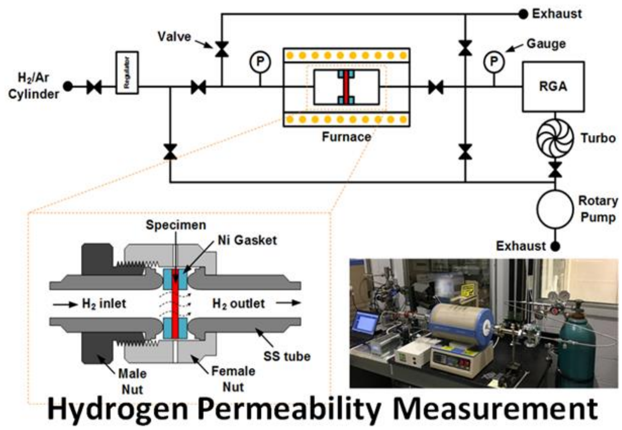


Figure 15. Hydrogen permeation measurement device established at Argonne National Laboratory.

A series of coated and uncoated Nb disk specimens were measured using this device. Preliminary results indicated that the permeation rate can be reduced by at least two orders of magnitude by adopting the metal-ceramic multilayer coating at 500°C. Further systematic measurement is ongoing.

5.1.4 Barrier coating to inhibit CMC diffusion within refractory metal liner

The approach for developing the CMC (SiC-based matrix) barrier layer has been very similar to the hydrogen permeation barrier design. Both standalone ceramic single layer and ceramic/ceramic and metal/ceramic based multilayer coating has been developed with selected materials to show the best diffusion barrier properties against SiC diffusion. Further systematic diffusion barrier studies are ongoing to identify the most optimized coating design against SiC (CMC).

5.1.5 Radiation tolerance of ALD multilayer barrier coating

The radiation tolerance of the barrier coating needs to be tested using swift ion irradiation at this stage. Both bulk specimen ion irradiation and *in situ* TEM ion irradiation experiments are planned. The bulk irradiation will be performed at the material irradiation chamber at Argonne Tandem Linac Accelerator System (ATLAS) [42] using 80 MeV Nb self-ions to 100 dpa (displacements per atom) at up to 900°C. The *in situ* TEM ion irradiation will be performed at Argonne's IVEM-Tandem facility using 1 MeV Kr ions at up to 800°C. The ion irradiation results will help to further improve the AMM design and manufacturing parameters to provide optimal radiation tolerance, as well as facilitate the design of future in-pile neutron irradiation campaign for AMM.

5.2 Miniature AMM Demonstration Plan

To demonstrate the concept of AMM and enhance the technology readiness level (TRL) and manufacturing readiness level (MRL), a miniature AMM will be assembled. The miniature AMM will be approximately 10 mm in diameter and 10 cm in length. The detailed plan for AMM assembly is listed as follows:

- <u>Metal hydride core</u>

The moderator core adopted for the miniature AMM will be yttrium hydride (YH_{2-x}) pellets. The pellets can either be made by hydriding machined yttrium metal or directly sintering yttrium hydride powder. The yttrium hydride pellets can also potentially be obtained from other national laboratories through Micro-Reactor Campaign or Transformational Challenge Reactor (TCR) program.

- <u>Coated refractory liner</u>

  At this stage, a Nb (or Mo) alloy thin-wall tube will be used as the metal liner material. Both the inner and outer surfaces of the tube will be coated by ALD multilayer barrier layers. The end caps will also be made of Nb and e-beam welded with the liner.

- <u>CMC cladding</u>

  The metal liner encapsulating the hydride core will be braided by SiC fiber (SiC_f) with pyrolytic carbon (PyC) interlayer coating. Then the braided SiC fiber will be infiltrated with SiC to form CMC cladding so that a miniature AMM with all essential components can be made.

The fabricated miniature AMM will be tested at elevated temperatures to demonstrate that yttrium hydride core can maintain its hydrogen density under such conditions.

6 Summary and Discussion

Advanced Moderator Modules (AMM) are being developed to boost neutron moderation in high-temperature micro-core concepts enabling improved core performance. The AMM concept can adopt a variety of hydride, CMC, refractory metal, and barrier coating materials for different types of micro-reactors and different envelopes of operating conditions. In this report, an AMM design that employs YH_{2-x} as moderating material surrounded by Nb-lined SiC/SiC cladding with advanced hydrogen barrier and metal-CMC diffusion barrier coating layers is introduced as a representative example. The structure of the AMM concept is provided in Section 3 with the functionalities of each component.

A preliminary assessment of the benefits of an AMM was completed in Section 4 based on a comprehensive multi-phase reactor physics analysis. The advanced enclosure solution proposed for the AMM would reduce U-235 enrichment by 6-8% when compared to a conventional enclosure solution (based on TZM alloy), for a micro-reactor based on the EMPIRE design fueled with TRISO. For the Holos Quad concept, employing the AMM technology enables either a significantly increased core life time or reduced core weight by 30-50%. The neutronic simulations demonstrate that AMM could be a game-changing concept to enhance the competitiveness of thermal neutron micro-reactors.

Ongoing demonstration efforts on the AMM concept were detailed in Section 5. These efforts are focused on the performance examinations of the key AMM component: advanced barrier coating, and its compatibility with metal liner under micro-reactor operation conditions. The preliminary results were reported. The advanced barrier coating was proven to provide satisfactory thermal cycle resistance under micro-reactor operating conditions. The excellent hydrogen barrier performance of the advanced barrier coating was also demonstrated. A more systematic development plan, including the fabrication of miniature AMM, was presented.

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